

Utilization of Orange Peel (*Citrus reticulata*) and Mussel Shells (*Perna viridis*) Waste as Polymer Reinforcers for Brake Lining Materials

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Article Information	ABSTRACT
Article History Received: November 17, 2025 Revised: December 05, 2025 Published: January 04, 2026 Keywords: <i>Brake pad;</i> <i>Citrus reticulata;</i> <i>Composite;</i> <i>Perna viridis;</i> <i>Waste paper.</i>	Environmentally friendly brake lining material uses a polymer composite reinforced with natural materials. Material testing of morphology, density, specific gravity, and swelling showed that composition significantly affects composite properties. Increasing the proportion of green mussel shell resulted in a rougher surface, higher density and specific gravity, and a significant decrease in swelling. The variation of paper, citrus reticulata, and perna viridis 0:50:50 had the highest degree of swelling (33.76%), while the variation of 75:12.5:12.5 showed the lowest value (0.17%), making it ideal for humid environments. The combination of orange peel and green mussel shell produces a lightweight, strong, and moisture-resistant material, making it an alternative, sustainable material for vehicle braking systems.

INTRODUCTION

The automotive industry is undergoing rapid development, driven by increasing demands for safer, more efficient, and environmentally sustainable products. A critical factor in ensuring driving safety is the proper functioning of the braking system. Brake pads, as essential components of the braking system, play a crucial role in reducing vehicle speed and preventing traffic accidents. With advancements in automotive technology, there is a growing need for brake pad materials that provide optimal braking performance while minimizing environmental impact (Awasthi & Saxena, 2019; Nandiyanto et al., 2022).

Composite materials consist of two or more distinct substances, each maintaining its individual characteristics, to form a new material with enhanced properties. For brake pad applications, employing polymers as a matrix reinforced with natural fibers represents a promising strategy. Natural fibers provide several benefits, including widespread availability, low production costs, and favorable mechanical properties (Arif et al., 2019).

Waste paper is the primary raw material for producing technical cardboard, which is used in roofing and car upholstery. Using waste paper with round mesh or folder machines reduces production costs, which is a key advantage (Wang et al., 2023). In contrast, specialized technical cardboard, such as filter cardboard, is manufactured from 50 to 100 percent organic and inorganic fibers, including basalt and cellulose derived from cotton fiber. These materials are typically formed into canvases, mats, or fabrics using specialized equipment and are subsequently impregnated or processed with specific resins to improve mechanical strength (Egamberdiev et al., 2024).

Orange peel (*Citrus reticulata*) represents a natural fiber with significant potential for composite development. This material constitutes a significant source of agricultural waste in numerous countries, particularly Indonesia, which ranks among the world's largest citrus

producers. Prior studies have demonstrated that orange peel fiber possesses a high cellulose content, rendering it suitable as a reinforcing material for composites (Marlina et al., 2021).

The shell of the green mussel (*Perna viridis*) demonstrates significant potential as a composite reinforcement material. This shell is abundant in calcium carbonate (CaCO_3), a compound recognized for its ability to enhance the mechanical strength of materials (Ayu Ariska et al., 2023; Wulansari et al., 2024). The presence of calcium carbonate in green mussel shells substantially enhances the tensile strength, stiffness, and resistance to abrasion and wear of polymer composites (Aridhani et al., 2021).

Using agricultural and marine waste as composite reinforcements supports zero-waste and sustainability principles (Liana et al., 2023). This concept highlights the need to minimize waste and use natural resources efficiently. Using agricultural waste like orange peels and marine waste such as mussel shells addresses waste issues while creating valuable new materials (Nastiti et al., 2024; Nika et al., 2020). Orange peel particles are low-density materials that easily fill large molds. As particle size increases, initial particle density rises, as seen with 100 and 140 mesh sizes, then stabilizes across different fiber percentages. The highest specific gravity is observed at 200 mesh. However, as fiber content increases from 10% to 30%, density decreases due to a higher percentage of voids.

Research on environmentally friendly composite materials has grown significantly. Using orange peel as a reinforcing agent in epoxy composites has notably improved mechanical properties, particularly tensile strength and hardness (Nurudin & Atmodjo, 2011). This study did not assess predictors of length of stay by discharge type, such as transfer to a ward or mortality. However, the data provide a reliable reference for future research (Desiasni et al., 2022). Combining orange peel and green mussel shells in a polymer composite is expected to yield brake lining material with strong braking performance, improved safety, and greater environmental sustainability (Pramono & Suprihanto, 2023).

The incorporation of shell powder as a reinforcing agent in polymer composite materials increases compressive and flexural strength. However, this addition reduces tensile strength. Specifically, the tensile strengths for composites containing 10% and 50% shell powder were 12.56 MPa and 9.18 MPa (Zulfikar et al., 2023). Natural composite materials offer not only sustainability but also economic benefits. The use of inexpensive, widely available orange peel and mussel shells reduces brake pad production costs (Arif et al., 2019; Ayu Ariska et al., 2023).

Natural fiber-based composite materials are lighter than metal-based alternatives. Consequently, polymer composites reinforced with natural fibers such as orange peel and mussel shells enhance braking performance and contribute to improved vehicle energy efficiency (Zulfikar et al., 2023). This research aims to develop a polymer composite brake lining reinforced with orange peel and green mussel shells. The study will assess the composite's morphology, density, and swelling.

RESEARCH METHODS

Materials and Instruments

The materials used in this study included distilled water, epoxy resin (R), hardener (H), green mussel shells (S), orange peels (O), waste paper (P), and thinner, each prepared according to laboratory-grade specifications. Prior to composite fabrication, all waste-based materials were manually cleaned, rinsed with distilled water, and dried in an oven at 100 °C for 1 hour to ensure moisture removal and surface cleanliness (Nandiyanto et al., 2022). The dried materials were then mixed with the resin–hardener system at a 40:60 ratio, using a mechanical stirrer to ensure uniform dispersion. The mixture was poured into molds and allowed to cure under ambient conditions. After curing, samples were demolded and prepared for characterization (Awasthi & Saxena, 2019).

Testing procedures included morphological analysis, density, and specific gravity measurements. Density and specific gravity were determined using the displacement method per ASTM D792 – Standard Test Methods for Density and Specific Gravity (Relative Density) of Plastics by Displacement, in which each sample was weighed in air and subsequently in distilled water to calculate the apparent density using Archimedes' principle. Specific gravity was determined by comparing the sample density with that of water at the same temperature, with

immersion time, medium temperature, and buoyancy correction controlled according to the standard to ensure accuracy and repeatability.

Swelling behavior was evaluated using the Flory–Rehner theory. Samples were immersed in solvent until equilibrium swelling was achieved, then weighed before and after immersion, and swelling values were calculated based on the mass increase following ASTM D570 procedures (Maleque et al., 2012). Additional details of the preparation, fabrication, and testing workflow are summarized in the flowchart provided in Figure 1.

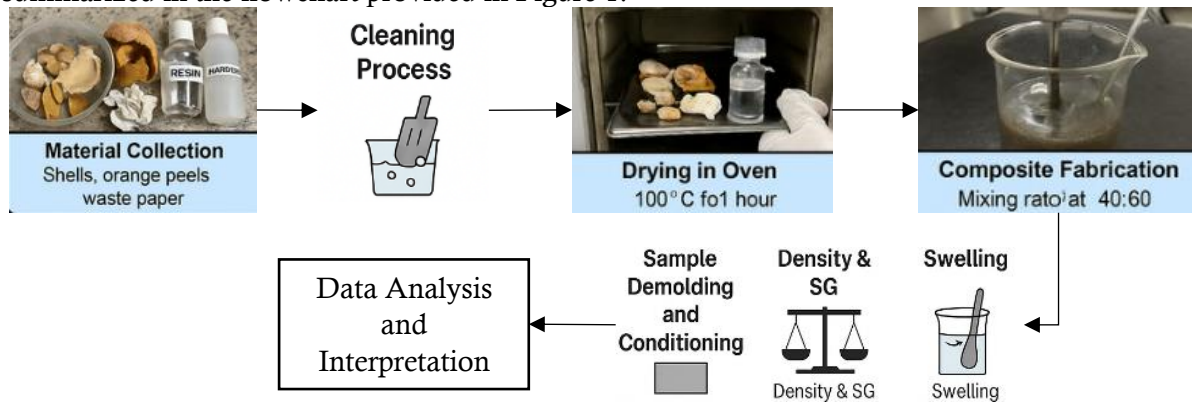


Figure 1. Composite surface testing for mass percentage P/O/S (%wt)

Analysis

The model was simplified to a two-group linear Scheffé model, which yielded the relevant equations from Equations 1 to 3 (Hao et al., 2021). The specific interaction effect between O and S cannot be evaluated due to collinearity, which limits the ability to test for deviations from linearity and to capture interaction effects (Kyriazos & Poga, 2023).

$$\rho_{POS} = \sum_{i=1}^3 \alpha_i \cdot x_i + \sum_{i < j} \alpha_{ij} \cdot x_i \cdot x_j \quad (1)$$

$$\text{Process limit } \sum x_i = 1, x_i \in [0,1] \quad (2)$$

$$\rho_{POS} = \alpha_1 \cdot x_P + \alpha_2 x_O + \alpha_2 x_S = \alpha_1 \cdot x_P + \alpha_2 (x_O + x_S) \quad (3)$$

$$SG_{POS} = \sum_{i=1}^3 \beta_i \cdot x_i + \sum_{i < j} \beta_{ij} \cdot x_i \cdot x_j \quad (4)$$

$$\text{Process limit } \sum x_i = 1, x_i \in [0,1] \quad (5)$$

$$SG_{POS} = \beta_1 \cdot x_P + \beta_2 x_O + \beta_2 x_S = \beta_1 \cdot x_P + \beta_2 (x_O + x_S) \quad (6)$$

The specific gravity (SG) dynamics within the P/O/S system were analysed using a mixture modelling framework based on Equations 4-6 (Hao et al., 2021). The degree of swelling was determined through nonlinear regression. Swelling data were first converted to the polymer volume fraction in the gel (ϕ), then fitted to obtain the interaction parameter (χ) and the crosslink density (v_e). The governing equation is as follows. (Richbourg & Peppas, 2020):

$$Q_m = \frac{m_{s120} - m_{s0}}{m_{s0}} \quad (7)$$

$$Q_v = 1 + \frac{\rho_p}{\rho_s} Q_m \quad (8)$$

$$\phi = \frac{1}{Q_v} \quad (9)$$

$$-[\ln(1 - \phi) + \phi + \chi \phi^2] = v_e V_1 \left(\phi^{\frac{1}{3}} - \frac{\phi}{2} \right) \quad (10)$$

Where Q_m is the mass swelling, derived from the swollen mass at 120 minutes (m_{s120}) and the initial dry mass (m_{s0}); denote the sample mass at 0 and 120 minutes, respectively. ρ_p and ρ_s represent the densities of the polymer and the solvent, respectively. ϕ is the polymer volume fraction in the gel. For water as the solvent, the molar volume is taken as ($V_1 = 18 \text{ cm}^3 \text{ mol}^{-1}$). The

column (Y_0) is a zero-valued response used in user-defined nonlinear regression to minimise the function $f(\phi; \chi, ve)$.

RESULTS AND DISCUSSION

Influence of Surface Testing

The surface morphology of two samples with different composition ratios, specifically 50:25:25 and 25:37.5:37.5, is depicted in Figure 2. The sample with a 50:25:25 composition exhibits a denser, more homogeneous surface and a deep orange color, suggesting an even distribution of filler within the polymer matrix. These observations suggest that a higher matrix proportion enhances the distribution of filler particles and strengthens the interphase bonds, resulting in a more uniform and compact structure (Dairi et al., 2017).

At a composition of 25:37.5:37.5, the surface exhibited increased roughness and heterogeneity, with coloration ranging from light yellow to golden. This observation suggests that higher filler content promotes particle agglomeration and imperfect dispersion, thereby reducing structural cohesiveness and increasing surface porosity. Such conditions are common in composite systems with elevated filler ratios, where limited interfacial interactions between polymer and filler phases decrease cohesion and increase the likelihood of microcracking during solidification. (Tazibt et al., 2023).

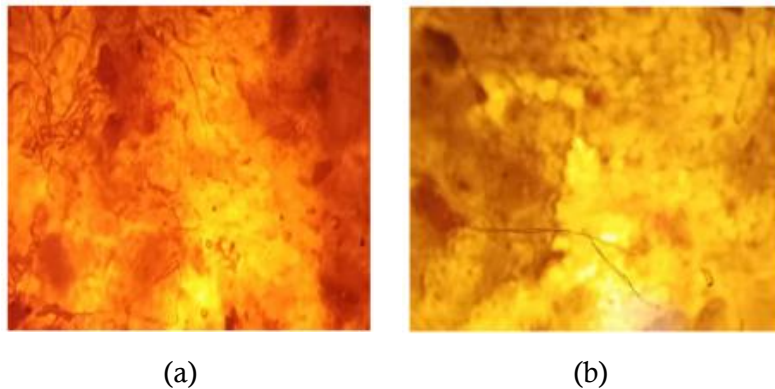


Figure 2. Composite surface testing for mass percentage P/O/S (a) 50:25:25 and (b) 25:37,5:37,5 (%wt)

An increase in the filler proportion from 25% to 37.5% reduces morphological homogeneity and adversely affects the mechanical and optical properties of the composite. These results align with previous findings, indicating that the optimum filler-matrix composition depends on interphase compatibility and mixing process conditions (Cionita et al., 2022).

Effect of Density and Specific Gravity

Density and specific gravity are critical parameters in the analysis of composite materials. Density refers to the mass per unit volume (g/L), whereas specific gravity represents the ratio of a material's density to that of water under specified conditions. According to Figure 3, the density values for the P/O/S variations of 0:50:50, 25:37.5:37.5, 50:25:25, and 75:12.5:12.5 are 1031.4 g/L, 1161.6 g/L, 1128.8 g/L, and 1235.6 g/L, respectively. The corresponding specific gravity values are 1,034.426 g/L, 1,165.028 g/L, 1,129.103 g/L, and 1,239.225 g/L. For P/O/S composites, density (mass per volume, g/L) and specific gravity (SG; the ratio of material density to water density) were analysed using a mixture model. From Equation 11, the variation in density and SG along the tested composition trajectory is primarily attributed to the increase in P fraction.

$$\rho_{POS} = 1282.8 x_P + 1052.1 x_O + 1052.1 x_S \quad (11)$$

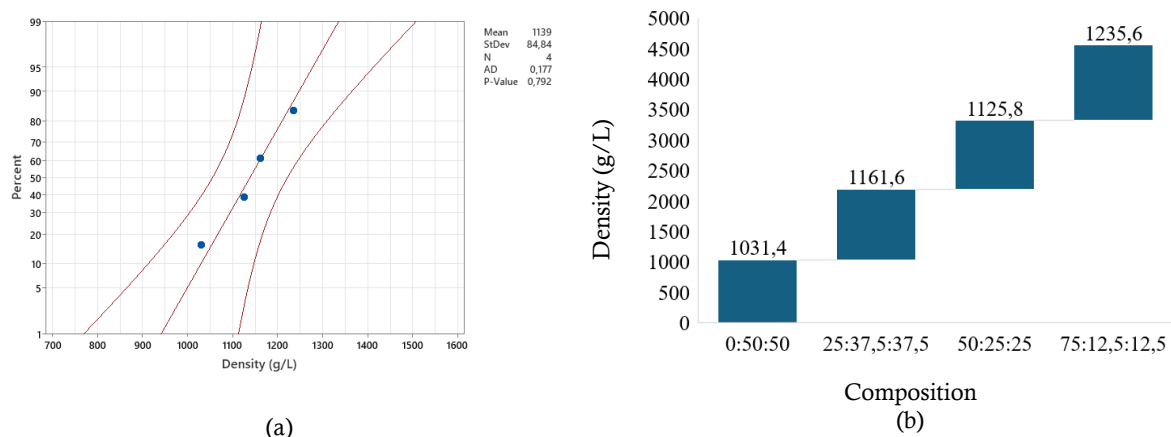


Figure 3. Influence of Density on the Mass Percentage of P/O/S (a) Probability Plot and (b) Density Chart

From Figure 4, at a 0:50:50 ratio, the lowest density and specific gravity indicate the predominance of lightweight materials, including orange peel and other composites, which are suitable for low-weight applications. Increasing the paper proportion to 25:37.5:37.5 results in higher density and specific gravity, indicating a denser material with enhanced mechanical strength.

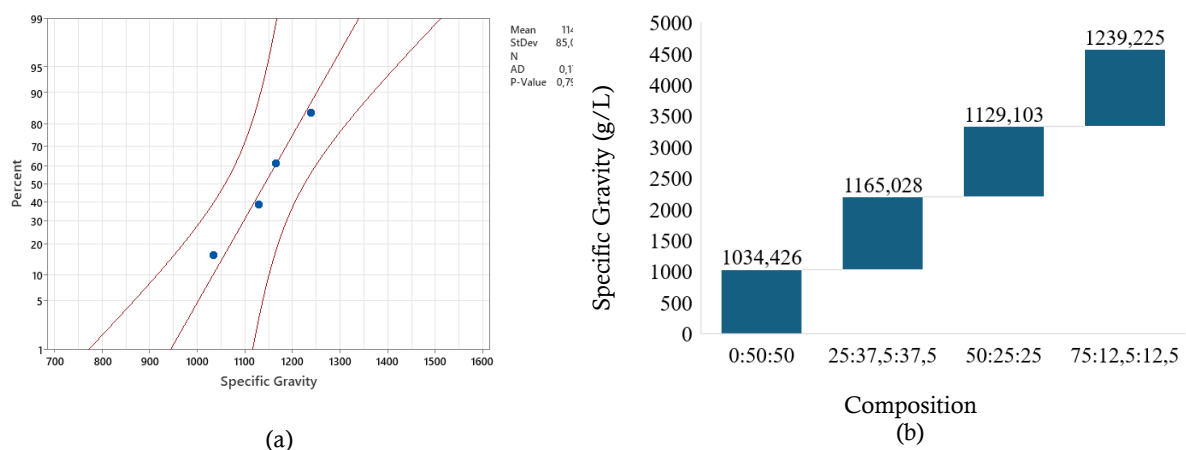


Figure 4. Influence Specific Gravity on the Mass Percentage of P/O/S (a) Probability Plot and (b) Specific Gravity Chart

Figure 4 provides both statistical and visual evidence to support these results. The probability plot indicates that the SG data are approximately normally distributed, as evidenced by a p-value of 0.7, which exceeds the 0.05 threshold and suggests no deviation from normality. The standard deviation remains approximately 85 g/L, consistent despite the small sample size ($n = 4$). The waterfall diagram demonstrates that SG increases with higher waste paper content, progressing from 0:50:50 to 75:12.5:12.5 (approximately 1034.426, 1165.028, 1129.103, and 1239.225 g/L). A minor decrease at 50:25:25 falls within the error margin and is likely attributable to mixture or packing heterogeneity. Density and specific gravity are closely related; specific gravity offers a relative measure compared to water, whereas density provides an absolute value. The observed slightly higher specific gravity results from the standard conversion to water density. Overall, these results indicate that increasing the proportion of paper enhances both density and specific gravity, highlighting the significant influence of composition on material performance.

Effect of Swelling Degree

The Flory–Rehner fit, based on Equation 12, terminated after 200 iterations, with parameter estimates constrained near their upper bounds ($\chi = 1.00$ and $v_e = 0.01$) and substantial standard errors. In conjunction with $DFE = 2$, $SSE = 24.37$, and $S = 3.49$, these results indicate poor

parameter identifiability and significant residual misfit. A formal lack-of-fit test could not be performed because replicate measurements were absent (Lawrance, 1994). The most plausible causes are a narrow span of polymer volume fractions (ϕ), too few data points, and potential inconsistencies in unit handling or conversion from mass swelling (Q_m) to ϕ . Consequently, the current parameter values should not be interpreted. To obtain meaningful estimates, future work should (i) collect at least 8–10 equilibrium ϕ points distributed across 0.10–0.60, (ii) verify ϕ conversions and unit consistency, and (iii) use reasonable initial guesses and parameter limits. If convergence remains difficult, fix χ to a literature value and estimate v_e only, or apply the linearised Flory–Rehner approach to determine the slope $v_e V_1$ (Friesen et al., 2022; Okumura et al., 2020).

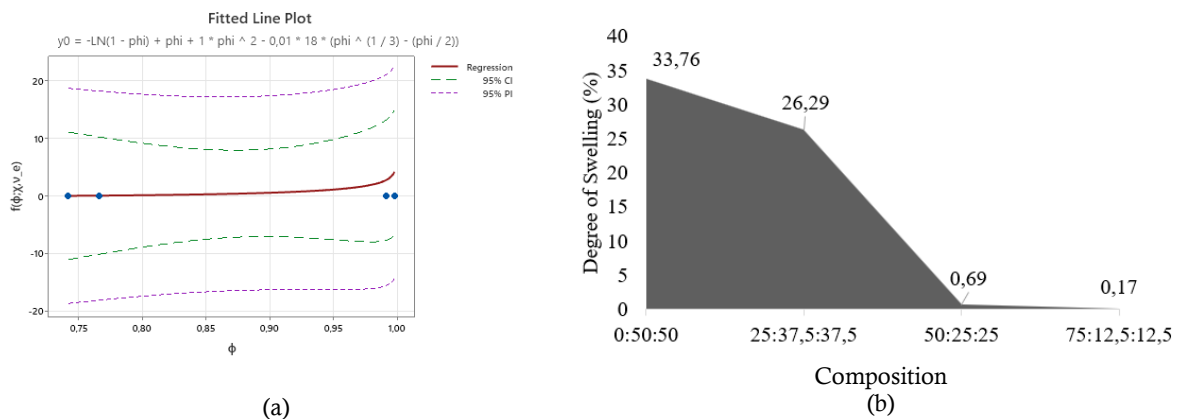


Figure 5. Influence of The Degree of Swelling at 120 min (a) Fitting Plot and (b) Degree Of Swelling Chart

Figure 5 presents two graphs depicting the relationships between material formulation variables and corresponding changes in physical properties. The regression graph on the left demonstrates that the function $f(\phi)$ remains relatively constant across variations in ϕ , as indicated by the nearly flat regression line. The widening of the confidence and prediction intervals at the extremes suggests limited data and increased model uncertainty in these regions. In contrast, the graph on the right shows a pronounced decrease in swelling as the composition changes, from 33.76% at the initial composition to 0.17% at a specific proportion. This substantial reduction suggests that the material becomes denser, more cross-linked, or more crystalline, resulting in a markedly lower liquid absorption capacity. The resulting fit, given in Equation 12, allows the coefficients to be interpreted as estimates of SG for the pure components. The analysis confirms that increasing proportions of P predominantly influence variations in SG along the tested compositional trajectory.

$$SG_{POS} = 1286.6 x_P + 1055.2 x_O + 1055.2 x_S \quad (12)$$

Figure 5 indicates that the swelling for the 0:50:50 variation is 33.76%, for 25:37.5:37.5 is 26.29%, for 50:25:25 is 0.69%, and for 75:12.5:12.5 is 0.17%. The highest swelling value is observed in the 0:50:50 mixture, attributable to the dominance of orange peel and other hydrophilic components, which facilitate greater water absorption. The swelling percentage decreases with the addition of waste paper, likely due to increased interfacial bonding at higher waste paper weight percentages. (Adediran et al., 2021). As the proportion of shells increases in the 75:12.5:12.5 hydrophobic variation, the swelling value decreases further. Materials with high swelling ratios, such as 0:50:50, are more prone to mechanical degradation due to water absorption. These materials are therefore less appropriate for applications that demand dimensional stability. In contrast, low-swelling compositions, such as 50:25:25 and 75:12.5:12.5, exhibit greater stability in wet environments, making them more suitable for applications requiring moisture resistance.

CONCLUSION

The automotive industry increasingly demands brake pad materials that are safer and more environmentally sustainable. Polymer composites reinforced with natural fibers offer an attractive option due to their affordability, wide availability, and favorable mechanical characteristics. Orange peel, which contains high levels of cellulose, and green mussel shells, which are rich in calcium carbonate, present strong potential as reinforcement agents that enhance strength, durability, and sustainability. Using agricultural and marine waste also aligns well with zero-waste principles. This study develops a polymer composite brake lining based on orange peel and mussel shells, focusing on morphology, density, and swelling, and proposes an environmentally friendly alternative to conventional brake pad materials.

Morphological observations reveal that a composition dominated by the polymer matrix produces a denser, more uniform surface. In contrast, the composition with a higher filler content exhibits rougher, more heterogeneous features due to particle agglomeration. Increasing filler content generally reduces structural uniformity and negatively affects visual and mechanical characteristics. Density measurements indicate that increasing the paper proportion results in a more compact composite structure, whereas lower paper content yields lighter materials characterized by greater orange peel and shell content. Swelling tests indicate a significant decline in water uptake as the proportion of paper and shell material increases, demonstrating improved dimensional stability and greater moisture resistance. These findings confirm that formulation strongly influences the composite's structural behavior and performance.

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